V. *On the production of regular double refraction in the molecules of bodies by simple pressure; with observations on the origin of the doubly refracting structure.* By DAVID BREWSTER, *LL.D. F.R.S. L. & E.*

Read February 11, 1830.

IN various papers already printed in the Philosophical Transactions, I have had occasion to show that the phenomena of double refraction may be produced artificially by certain changes in the mechanical condition of hard and soft solids[*]. In all these cases the phenomena are related to the form of the mass in which the change is induced; and in the case of hard and elastic solids, they vary with any variation of form which alters the mechanical state of the particles. In isinglass and other bodies to which double refraction has been communicated by induration, the particles take a permanent position, which is not altered by any change of shape; but still the phenomena exhibited by a given portion of the mass are related to the surfaces where the indurating cause operated, and also to those by which the isinglass was bounded; and they depend on the position which that portion occupies in the general mass.

In all these cases the phenomena are entirely different from those of regular crystals, and in none of them is the doubly refracting force a function of the angle which the incident ray forms with one or more axes given in position.

As long ago as 1814 I communicated to the Royal Society the following experiment on the depolarizing structure of white wax and resin:

" When resin is mixed with an equal part of white wax, and is pressed between two plates of glass by the heat of the hand, the film is almost perfectly transparent by transmitted light, though of a milky white appearance by reflected light. It has not the property of depolarization when the light is incident vertically; but it possesses it in a very perfect manner at an oblique incidence, and exhibits the segments of coloured rings[†]".

[*] Phil. Trans. 1814; 1815, pp. 1, 30, 60; 1816, pp. 46, 56.
[†] Ibid. 1815, pp. 31, 32.

The subject of double refraction was then so little developed that this expe-
riment excited no notice; and it was only brought to my own recollection by
the accidental appearance of the specimen itself. This depolarizing film has
suffered no change by remaining fifteen years between the plates of glass. The
vertical line along which it is destitute of the property of depolarization is a
single axis of double refraction; and the coloured rings at oblique incidences
are produced by the inclination of the refracted ray to the axis of double re-
fraction. In order to examine this remarkable effect under a more general
aspect, I made a considerable number of such plates with different kinds of
wax, and with various proportions of resin, and I was led to results which
seem to possess considerable interest.

When the white wax is melted alone and cooled between two plates of glass,
it consists of a number of minute particles, each possessing double refraction,
but having their axes turned in all possible directions. If the film of wax is
made extremely thin, the particles are not sufficiently numerous to exhibit any
action upon polarized light.

When resin alone is melted and cooled in a similar manner, it exhibits no
doubly refracting structure, whether it indurates slowly or under the influence
of pressure.

If resin and white wax are mixed in nearly equal proportions, the compound
possesses considerable tenacity. When a proportion of it is melted and cooled
between two plates of glass, it shows the quaquaversus polarization of bees'-
wax, the axes of the elementary particles being turned in every direction. It
possesses a considerable degree of opalescence, and a luminous body seen
through it is surrounded with nebulous light. This imperfect transparency
evidently arises from the reflexion and refraction of the rays in passing from
one molecule to another, occasioned by a difference in the refractive power of
the ingredients, or by the imperfect contact of the particles, or by both these
causes combined.

In order to observe the modifications which these phenomena received from
pressure, I took a few drops of the melted compound and placed them in suc-
cession on a plate of thick glass, so as to form a large drop.   Before it was
cold, I laid above the drop a circular piece of glass about two thirds of an inch
in diameter, and by a strong vertical pressure on the centre of the piece of

glass, I squeezed out the drop into a thin plate. This plate was now almost perfectly transparent, as if the pressure had brought the particles of the substance into optical contact.

If we expose this plate to polarized light, we shall find that it possesses one axis of positive double refraction, and exhibits the polarized tints as perfectly as many crystals of the mineral kingdom. The structure thus communicated to the soft film by pressure does not belong to it as a whole, nor has it only one axis passing through its centre like a circular piece of unannealed glass. In every point of it there is an axis of double refraction perpendicular to the film, and the doubly refracting force varies with the inclination of the incident ray to this axis, as in all regular uniaxal crystals. When the two plates of glass are drawn asunder, we can remove one or more portions of the compressed film, and these portions act upon light exactly like films of uniaxal mica or hydrate of magnesia, and develope a doubly refracting force of equal intensity.

This remarkable experiment presents an interesting subject of inquiry. That the regular double refraction of the film is developed by the agency of pressure cannot be doubted; but it does not at first sight appear whether it is the immediate effect of the pressure, or is the same doubly refracting force which produces the quaquaversus polarization that takes place when the resinous film indurates without constraint. In this state of the film the axes of double refraction are clearly turned in every conceivable direction; and it is impossible to suppose that a pressure in one direction could suddenly arrange all these axes in parallel positions. The double refraction of each particle of the film has therefore been developed by the compressing force similarly applied to them; and in producing this effect, it must have deprived each particle of the doubly refracting structure which it previously possessed. The substitution of one doubly refracting structure for another may be easily effected in many bodies. Even in regular crystals we can by heat or pressure modify or remove their double refraction. Nay, we can take away one axis from a biaxal crystal, and communicate a second axis to an uniaxal one. When the doubly refracting structure is produced by induration, we can remove it wholly by pressure, and replace it with another even of an opposite character; and when it is generated by the living principle, as in the case of

the crystalline lenses of animals, we can take it away entirely, and substitute a new and more powerful doubly refracting structure by induration.

We may therefore consider it as clearly established that the uniaxal double refraction of the resinous mass has been communicated to the individual molecules by simple pressure; the increased transparency arising from the molecules being brought into closer contact, and the regular double refraction from the variable density impressed upon each elastic molecule, and symmetrically related to the axis of pressure. The effect thus produced on the resinous mass is precisely the same as what would take place by subjecting elastic spheres to a regular compressing force. The axis of pressure becomes an axis of positive double refraction, and the double refraction increases with the inclination of the ray to the axis, and becomes a maximum in the equator of the molecules.

By this view of the preceding facts, we are led to a very simple explanation of the origin and general phenomena of double refraction in regular crystals. That this property is not inherent in the molecules themselves may be easily proved. The particles of silex, for example, do not possess it in their separate state. In tabasheer, in many opals, and in melted quartz, there is not the slightest trace of the doubly refracting structure: but when the particles of silex in solution are allowed to combine, in virtue of their polarities or mutual affinities, they then instantly acquire, at the moment of their combination, the property of double refraction, and they retain it while they continue in this state of aggregation. The manner in which this takes place may be easily conceived: a number of elastic molecules existing in a state of solution, or in a state of fusion, are kept at such a distance by the fluid in the one case, and by the heat in the other, as to preclude the operation of their mutual affinities; but when, in the process of evaporation or cooling, any two molecules are brought together by the forces or polarities which produce a crystalline arrangement, and strongly adhere, they will mutually compress one another, and each will have an axis of double refraction in the directions of the line joining their centres, in the same manner as if they had been compressed by an external force.

From the phenomena of crystallization and cleavage, it is obvious that the molecules of crystals have several axes of attraction, or lines along which they

are most powerfully attracted, and in the direction of which they cohere with different degrees of force. Guided by the indications of hemitrope forms, and supposing the molecules to be spherical or spheroidal, we infer that their axes are three in number and at right angles to each other, and are related in position to the geometrical axis of the primitive form. In like manner the phenomena of double refraction are related to the same axis of the primitive form, and may be all rigorously calculated by a reference to three rectangular axes. In uniaxal crystals, the three axes A, B, C must be such that two of them are equal and of the same name; while the third, corresponding with the apparent axis, may be of the same or of a different name. In biaxal crystals, the three axes A, B, C are unequal, and in crystals with no double refraction the axes are equal and destroy each other*.

This approximation of these two classes of facts is too remarkable to be accidental, and would go far to establish their dependence, even if it were not indicated by other arguments which I shall proceed to illustrate.

Among those crystals which have the obtuse rhomboid for their primitive form, there are many with one axis of negative double refraction, and only one or two with one axis of positive double refraction. In the former, the negative doubly refracting structure will be produced round the axis of the rhombohedron by the compression arising from attractions in the direction of two equal rectangular axes A, B, which will dilate the molecules in the direction of the third axis C, and make it a negative axis of double refraction, equal in intensity to either of the other two. Here we require the combination only of two axes; but if we suppose that there is in the direction of C a third axis of attraction either more or less powerful than the other two, then if it is less powerful, the compression of the molecules produced by it will diminish the dilatation arising from the united action of A and B, but will still leave an unbalanced dilatation, or a single negative axis of double refraction in the axis of the rhomb.

If C, on the contrary, is an axis in which the attractive force of the mole-

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* In uniaxal crystals, the resultant of the two equal axes A, B may have any relation to C but that of equality; excepting when C is of a different name from A and B.

In biaxal crystals, any two axes A, B, may be converted into three A + C, B ± C, ± C. See Phil. Trans. 1818.

cules is greater than along A and B, the compression which it produces will exceed the dilatation arising from A and B, and we shall have an axis of compression along C, or an axis of positive double refraction as in quartz and dioptase*. The same observations are applicable to minerals that crystallize in the pyramidal form.

When the three axes A, B, C are all equal, the three rectangular compressions, produced by the aggregation of the molecules, will destroy one another at every point of the molecule, and the body which they compose will have no double refraction, and cleavages of equal facility. Hence all crystals in which it is known by cleavage that the particles cohere with equal force in three rectangular directions have actually no double refraction.

If the three attractive axes A, B, C are all unequal, the difference of density which they produce in the molecules will be related to two axes of double refraction, the strongest of which will be negative or positive according as the compression along C is less or greater than the dilatation produced along C by the united compressions of A and B. Hence all crystals belonging to the prismatic system, in which we are informed by cleavage that the particles cohere with unequal forces in three directions, have invariably two, or, as we have already explained, three unequal axes of double refraction, of which the strongest is sometimes positive and sometimes negative.

We have supposed the elementary molecules of bodies to be spherical when existing singly, or beyond the sphere of their mutual action; but although their form must, in the case of doubly refracting crystals, be changed into oblate, prolate or compound spheroids, yet the deviation of these spheroids from the sphere may be so small, that the forms of the bodies which they compose may be regarded as arising from the union of spherical molecules. It is more probable, however, that the form of the molecules suffers a considerable change, and we may consider that change as determining the exact primitive form of the crystal and the inclination of its planes.

The circumstance of almost all rhombohedral crystals having negative

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* Since this paper was written, I have seen the very valuable researches of M. SAVART on the structure of crystallized bodies as developed by sonorous vibrations. The curious result of his experiments, that the axis of calcareous spar, a negative axis of double refraction, is the axis of least elasticity, while the axis of quartz, an axis of positive double refraction, is the axis of greatest elasticity, harmonizes in a remarkable manner with the above views.

double refraction, which can only be produced by axes of compression in the equator of a prolate spheroid, excludes the supposition, that the ultimate mole- cules are spherical particles converted by the forces which unite them into those oblate and prolate spheroids, by means of which, according to the views of HUYGENS, all the varieties of rhombohedrons may be formed*; for if this were the case, the obtuse rhombohedrons should possess one positive axis, and the acute ones one negative axis of double refraction. We are constrained therefore to suppose that in rhombohedral crystals the molecules have the form of an oblate spheroid, with its axes so related, that the change superin- duced upon it by the forces of aggregation determines the exact form of the combination. In carbonate of lime for example, where the precise inclination of the faces of the rhombohedron can be produced only by oblate spheroids whose polar is to their equatorial axis as 1 to 2.8204, we may suppose that the spheroids were originally more oblate, and that the forces by which they receive the doubly refracting structure dilated them in the direction of the smaller axis, so as to produce a spheroid having its axis as 1 to 2.8204. Hence if we could suppose the molecules placed together without any forces which would alter their form, they would compose a rhombohedron with a greater angle and having no double refraction. But when they are combined by the attractive forces of crystallization, they compose a rhombohedron of 105°, pos- sessing negative double refraction.

In this view of the subject, the form of the ultimate molecules of crystals existing separately, may be regarded as determining within certain limits the primitive form to which they belong; while the doubly refracting structure and the precise form of the crystal are simultaneously produced by the action of the forces of aggregation.

These views receive a remarkable illustration from a new doubly refracting structure, which I discovered many years ago in chabasie, and which will form the subject of a separate communication. In certain specimens of this mi- neral, the molecules compose a regular central crystal, developing the pheno- mena of regular double refraction; but in consequence of some change in the state of the solution, the molecules not only begin to form a hemitrope crystal

* See HUYGENS's Traité de la Lumière, chap. v. and the Edinburgh Journal of Science, No. xviii. pp. 311, 314.

on all the sides of the central nucleus, but each successive stratum has an in-
ferior doubly refracting force till it wholly disappears. Beyond this limit it
reappears with an opposite character, and gradually increases till the crystal is
complete. In this case the relative intensities of the axes or poles from which
the forces of aggregation emanate, have been gradually changed, probably by
the introduction of some minute matter, which chemical analysis may be un-
able to detect. If we suppose these axes to be three, and the foreign particles
to be introduced, so as to weaken the force of aggregation of the greater axis,
then the doubly refracting force will gradually diminish with the intensity of
this axis, till it disappears, when the three axes are reduced to equality. By
continuing to diminish the force of the third axis, the doubly refracting force
will reappear with an opposite character, exactly as it does in the chabasie un-
der consideration.

From the mutual dependence of the forces of aggregation and double refrac-
tion, it is easy to understand the influence which heat produces on the doubly
refracting structure, as exhibited in the phenomena discovered by M. MITS-
CHERLICH in sulphate of lime and calcareous spar, and in those which I detected
in glauberite*. This eminent philosopher has found, by direct experiment, that
heat expands a rhomb of calcareous spar in the direction of its axis, and con-
tracts it in directions at right angles to that axis†; that the rhomb thus be-
comes less obtuse, approaching to the cubical forms which have three equal axes,
and that its double refraction diminishes. All these effects are the necessary con-
sequences of the preceding views. The expansion in the direction of the axis,
and the contraction of all the equatorial diameters diminish the compression
of the axes of the oblate spheroidal molecules, and must therefore diminish its
double refraction, as well as the inclination of the faces of the rhomb. In

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* See Edinburgh Transactions, vol. xi.

† It follows from this fact, that massive carbonate of lime, in which the axes of the molecules have
every possible direction, should neither expand nor contract by heat, and would therefore form an
invariable pendulum. As there must be, in any given length of massive carbonate of lime, as many
expanding as there are contracting axes, then, if the contractions and expansions in each individual
crystal are equal, they will destroy one another ; but if they are proportional to their lengths, the con-
tractions will exceed the dilatations. In this case, we have only to combine the marble with an ordi-
nary expanding substance, to have an invariable pendulum. The balances of chronometers might
be thus made of mineral bodies.

like manner it will be found that in sulphate of lime and glauberite the expansions and contractions will be so related to the three axes, as to explain the conversion of the biaxal into the uniaxal structure, and the subsequent reappearance of the biaxal structure in a plane at right angles to that in which the axes are found at ordinary temperatures.

The phenomena exhibited by fluids under the influence of heat and pressure, and those of doubly refracting crystals, exposed to compressing or dilating forces, are in perfect conformity with the above views ; so that even without the fundamental experiment described in this paper, we might have been entitled to conclude that the forces of double refraction are not resident in the molecules themselves, but are the immediate result of those mechanical forces by which these molecules constitute solid bodies.

Allerly, October 5th, 1829.